where the subscript I refers to the initial ratio at time *T* and the subscript I, Earth refers to the initial ratio for the bulk earth at rock age *T*. We have used the initial ^{143}Nd/^{144}Nd value of the achondrite meteorite Juvinas [G. W. Lugmair, J. P. Kurtz, K. Marti, *Proc. 6th Lunar Sci. Conf.* (1975), p. 1419; (*9*)] and the averaged ^{147}Sm/^{144}Nd ratio of 0.1936 in chondrite meteorites [N. Nakamura, *Geochim. Cosmochim. Acta* **38**, 757 (1974)] as representative of a chondritic bulk earth of age 4560 × 10^6 years.

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Characterization of Plutonium in Maxey Flats Radioactive Trench Leachates

Abstract. *Plutonium in trench leachates at the Maxey Flats radioactive waste disposal site exists as dissolved species, primarily complexes of the tetravalent ion with strong organic ligands such as ethylenediaminetetraacetic acid. The complexes are not sorbed well by sediment and are only partly precipitated by ferric hydroxide. These results indicate the importance of isolating radioactive waste from organic matter.*

Concern over the possible migration of plutonium from radioactive waste repositories has led to a flurry of investigations to determine and predict plutonium's behavior in ground waters. Initial efforts were concentrated on the determination of distribution coefficients (erroneously referred to as K_d values, which imply that equilibrium is attained) between synthetic ground waters and various rock types. There has been lately a growing realization that for such a complex, multivalent element, distribution coefficient values are meaningless unless the plutonium species present in the system is identified. Consequently, speciation research is under way in several laboratories.

The U.S. Geological Survey has instituted a program to characterize plutonium in radioactive leachates from disposal sites in different geologic formations. We report here the results of research at the Maxey Flats radioactive waste disposal site in eastern Kentucky. This site received waste from research laboratories, hospitals, nuclear power stations, and commercial radioisotope users from 1963 until it was closed in 1977; it contains an estimated 80 kg of ^{239}Pu as well as significant quantities of ^{238}Pu (*1*). The solid waste, which was contained in steel, cardboard, wood, or plastic containers, was placed in trenches approximately 76 to 110 m long, 6 m deep, and 6 m wide (*2*). In addition, there were a few trenches (the 33-L series) in which liquid wastes were placed and subsequently solidified. After filling, the trenches were covered with at least 1 m of compacted soil.

Because the material in the filled trenches is less compacted than the surrounding soil, water collects in the trenches as a result of the high rainfall in the area (*3*). This water leaches material, including radioactive isotopes and a number of organic compounds, from the waste, and has to be removed to prevent overflowing the tops of the trenches. Consequently, standpipes were installed, and the malodorous, foul-tasting (*4*) water—referred to as leachate—is periodically pumped from the trenches and evaporated. It was through these standpipes that we obtained leachate samples for this study.

We obtained samples for this study during two different time periods. Trenches 2, 26, 32, 33-L-18, and 35 were sampled in October 1978 and trenches 19S, 23, 27, 33-L-4, 33-L-18, and 35 in November 1979. Two trenches, 33-L-18 and 35, were sampled in both periods to assess the variation in behavior with time. All trenches were sampled at least twice, using a fresh, clean filtration assembly for each sampling. The procedure consisted of filtering the leachate sequentially through a series of Nucle-

Table 1. Filtration results. Horizontal rows refer to separate sampling runs; numbers in parentheses are 1 standard deviation for four replicate analyses.

Trench	*p*H	^{238}Pu (pCi/liter)		^{238}Pu in filtrate (%)	^{239,240}Pu (pCi/liter)	
		Unfiltered	0.05-µm filtrate		Unfiltered	0.05-µm filtrate
2	6.9	1,180 (7)	1,170 (45)	99	225 (38)	225 (11)
	6.8	1,216 (36)	1,440 (45)	~100	243 (5)	293 (6)
		1,260 (63)	1,580 (11)	~100	288 (9)	315 (14)
19S	6.6	74,700 (500)	76,800 (1,500)	~100	560 (50)	<10
	6.6	82,400 (1,500)	87,200 (500)	~100	560 (50)	<10
23	7.0	36,500 (400)	35,200 (200)	96	<10	240 (50)
	7.0	36,300 (700)	34,800 (1,400)	96	<10	240 (50)
26	6.8	8.2(0.5) × 10^5	5.0(0.05) × 10^5	61	<10	<10
	7.0	4.5(0.05) × 10^5	3.0(0.3) × 10^5	67	<10	<10
	6.7	64,000 (9,000)	56,000 (1,000)	88	<10	<10
27	6.0	3,560 (200)	2,080 (200)	58	430 (50)	220 (50)
	6.2	9,130 (500)	3,900 (500)	43	860 (50)	320 (80)
32	7.8	20,300 (400)	24,900 (300)	~100	340 (30)	360 (20)
	7.5	26,400 (300)	24,600 (100)	93	410 (50)	230 (50)
	7.8	39,400 (300)	27,300 (100)	87	380 (20)	250 (10)
33-L-4	12.2	1,160 (350)	1,030 (320)	89	6,640 (110)	8,320 (380)
	11.8	1,050 (320)	970 (300)	92	7.910 (110)	7,480 (190)
33-L-18	1.9	10,000 (1,400)	8,400 (100)	84	3,000 (300)	2,200 (140)
	2.2	9,500 (900)	6,300 (200)	66	2,500 (200)	2,200 (90)
		4,640 (50)	3,620 (240)	78	1,460 (30)	1,210 (80)
35	8.6	12,900 (100)	11,200 (300)	87	200 (20)	90 (50)
	8.1	12,800 (50)	11,700 (90)	91	230 (30)	230 (50)
	8.4	4,750 (400)	4,370 (400)	92	<10	<10

pore membrane filters with pore sizes of 5, 0.4, and 0.05 μm and collecting a sample of filtrate from each filter. Samples of the unfiltered water and each filtrate were acidified to $1N$ with HNO_3.

In the case of the 0.05-μm filtrate, additional samples were collected in Teflon bottles under an argon atmosphere without acidification and subjected, within 30 minutes of collection, to a series of separations, in an inert atmosphere, to establish the charge and oxidation states of the plutonium. Specifically, separate 100-ml portions of the 0.05-μm filtrate were passed through anion exchange resin and cation exchange resin, after which the cation resin was eluted with $8M$ HNO_3 and the anion resin with $0.4M$ HNO_3 and $0.04M$ HF. The eluates and effluents were analyzed for plutonium to determine the fractions in anionic, cationic, and uncharged forms. To determine oxidation states, we subjected separate samples of the 0.05-μm filtrates to a series of carrier precipitation and solvent extraction procedures (*5*). Praseodymium fluoride precipitation, in the absence of a reducing agent, carries the reduced forms of plutonium (Pu^{3+} and Pu^{4+}); when precipitated after addition of a reducing agent, such as $NaHSO_3$, to reduce any oxidized plutonium (PuO_2^{+} and PuO_2^{2+}), PrF_3 carries, at least in theory, all the plutonium. The reduced forms were also carried by zirconium iodate precipitation, and the oxidized forms by precipitation of sodium uranyl acetate. Solvent extraction with $0.5M$ thenoyltrifluoroacetone (TTA) in xylene was used to separate plutonium (IV). Carrier precipitation samples were dissolved in $1M$ HNO_3 and TTA extracts were evaporated at room temperature before analysis.

For statistical purposes, four replicate portions of each solution were analyzed. The same basic procedure was followed for plutonium analyses of all samples and has been described elsewhere (*6*). To monitor recovery, ^{236}Pu (or ^{242}Pu) tracer was added to each sample before the initial organic destruction step. Counting periods were long enough to achieve satisfactory statistics—generally 1000 minutes. The yield of the overall procedure was calculated from the recovery of the tracer.

Of the nine trenches sampled, seven had received only solid waste, whereas two—33-L-4 and 33-L-18—had been filled with liquid waste. The liquid waste in these trenches was solidified with cement in 33-L-4 and by formation of a urea-formaldehyde resin ("Tiger-Lok") in 33-L-18.

Table 2. Plutonium speciation results. Only ^{238}Pu data are reported here; the data for $^{239,240}Pu$ are similar.

Trench	Plutonium (%) by ion exchange				Plutonium (%) by PrF_3 precipitation*			Reduced Pu in tetravalent state (%)§
	Anionic	Cationic	Uncharged	Material balance	Reduced Pu†	Oxidized Pu‡	Total Pu	
19S	41	13	38	92	77	8	85	100
	49	14	32	95	94	<1	94	76
23	41	19	41	101	90	4	94	79
	41	16	43	100	86	3	89	85
27	49	10	6	65	76	3	79	2
	62	6	21	89	78	<1	78	6
33-L-4	32	15	56	103	81	18	99	>100
	28	13	64	105	83	25	>100	>100
33-L-18	25	3			100	<1	100	91
35	71	18	<1	89	86	2	88	94
	41	12	35	88	81	<1	81	66

*In the PrF_3 precipitation data the percentage is relative to the total Pu present, as determined by direct analysis of the 0.05-μm filtrate. †Determined by direct PrF_3 precipitation of the 0.05-μm filtrate. ‡Determined by reduction of all Pu with $NaHSO_3$ followed by PrF_3 precipitation, minus the Pu value obtained in the precipitation without prior reduction. §Defined as the percentage of Pu extracted by TTA-xylene relative to the total reduced Pu concentration determined by PrF_3 carrier precipitation.

The leachates varied widely in their concentrations of phosphate, fluoride, and dissolved organic carbon. With the exception of leachate from trench 33-L-18, which had 2400 mg/liter, all had low concentrations of phosphate (< 1 mg/liter). The fluoride ion concentration was relatively high only in trench 35 leachate (35 mg/liter). The disposal of detergent solutions used for decontaminating equipment into trench 33-L-18 accounts for the high phosphate concentration in leachate from this trench. Concentrations of dissolved organic carbon were relatively high, especially in the leachates from 33-L-4 and 33-L-18 (1200 and 2100 mg/liter, respectively). Leachates in the solid-waste trenches were near neutrality, whereas the respective *p*H values of leachates from 33-L-4 and 33-L-18 were 12.0 and 2.0 as a result of the solidification agents used in them.

The unfiltered leachates and the 0.05-μm filtrates were analyzed with the results shown in Table 1. It should be noted that the standard deviations shown in Table 1 merely indicate the reproducibility of the determinations and do not take into account variations in concentrations in different samples resulting from inhomogeneity of the trench leachates. For example, in several instances the 0.05-μm filtrates actually had a higher plutonium concentration than the unfiltered trench leachates, and in the third sampling run of trench 26, made 2 days after the first two runs, the plutonium concentrations were less by about an order of magnitude.

The data in Table 1 show that in the leachate from all trenches except trench 27 (and, to a lesser extent, trench 26), most of the plutonium is in true solution or contains only very small colloidal particles; absence of colloids composed of high-molecular-weight polymers is clearly demonstrated. Some of the trench leachates contained appreciable amounts of suspended sediment that were removed by the filters. The fact that little if any of the plutonium was sorbed onto these sediments suggests that the plutonium is not colloidal, and this conclusion is further corroborated by data cited below. Only in the leachates from trenches 27 and 26 was there evidence for the presence of a significant fraction of plutonium in colloidal form. Other data (reported below) also indicate that leachate in trench 27 differs from that in the other solid-waste trenches sampled.

The exceptionally low solubility of plutonium in water suggests that its presence in these leachates is the result of complex formation, particularly in view of the relatively large concentrations of dissolved organic carbon. This presumption is supported by the ion exchange data in Table 2. A larger percentage of plutonium was sorbed by the anion exchange resin than by the cation resin in every instance. In leachate from some trenches—particularly 33-L-4—a large fraction of the plutonium existed in uncharged form, as determined by analysis of the anion and cation exchange effluents. The material balance of one of the trench 27 leachates was very low, suggesting that some of the plutonium in this water was irreversibly sorbed onto the anion or cation resin, or both. This fraction may be low-molecular-weight colloidal material.

It is most unlikely that the plutonium

Table 3. Ferric hydroxide precipitation results. Horizontal rows refer to separate sampling runs; numbers in parentheses are 1 standard deviation for four replicate analyses.

Trench	^{238}Pu (pCi/liter)		^{238}Pu remaining in solution (%)	^{239,240}Pu (pCi/liter)		EDTA (mg/liter)
	Initial 0.05-µm filtrate*	After Fe(OH)$_3$ precipitation		Initial 0.05-µm filtrate*	After Fe(OH)$_3$ precipitation	
2	1170 (50)	51 (14)	5	225 (11)	<10	
	1580 (10)	110 (5)	7	315 (14)	18 (4)	N.A.†
19S	$8.72(0.05) \times 10^4$	7500 (400)	9	<10	71 (5)	0.1
23	$3.65(0.04) \times 10^4$	$1.41(0.07) \times 10^4$	38	560 (50)	223 (15)	‡
26	$5.6(0.1) \times 10^4$	980 (70)	2	<10	<10	0.5
	$5.0(0.05) \times 10^5$	1850 (80)	<1	<10	<10	
27	3900 (500)	630 (110)	16	320 (80)	79 (30)	0.1
32	$2.46(0.01) \times 10^4$	8060 (200)	33	230 (50)	<10	0.2
33-L-4	1030 (320)	790 (400)§	77	8320 (380)	7390 (700)	0.1
33-L-18	6300 (200)	720 (80)	11	2200 (100)	<10	N.A.
	3620 (240)	3740 (50)§	100	1210 (80)	940 (10)	N.A.
35	$1.12(0.03) \times 10^4$	3500 (200)	31	90 (50)	58 (19)	0.1
	4370 (400)	1840 (330)	42	<10	160 (61)	N.A.

*See Table 1. †Not analyzed because of interferences in solution. ‡Considerable EDTA apparently present, but quantitative analysis not possible. §No precipitate.

is complexed by inorganic ligands such as fluoride, phosphate, or carbonate. If phosphate complexes had been present, they would have caused low yields in the analytical procedure. Carbonate and fluoride complexes would have been destroyed by the initial acid fuming. The fact that good analytical recovery of plutonium required drastic organic destruction measures further suggests that the plutonium is complexed by organic ligands.

Results of oxidation state determinations indicated that the plutonium in the leachate from every trench is almost entirely in the reduced form. The PrF$_3$ carrier precipitation data (Table 2) show that only in leachate from trench 33-L-4 is there an appreciable amount of higher oxidation states. This is corroborated by the Zr(IO$_3$)$_4$ and sodium uranyl acetate precipitation data, except that the latter results indicate that approximately one-third of the plutonium in trench 33-L-18 leachate is in the oxidized states, and that the total oxidized plus reduced plutonium in trench 27 leachate is considerably less than 50 percent of the plutonium present. This further anomalous behavior of trench 27 leachate is unexplained.

Extraction with TTA-xylene is specific for plutonium (IV) (*7*). The data for these extractions (Table 2) indicate that, except for leachate from trench 27, most of the reduced plutonium—and hence total plutonium—is in the tetravalent state. This finding is not surprising since plutonium (IV) is the most strongly complexed of the four common oxidation states (*7*, p. 92).

Archival samples of each 0.05-µm filtrate were taken under an inert atmosphere and returned to our laboratory for later study. The trench leachates contain appreciable concentrations of iron, and several months after collection it was found that an Fe(OH)$_3$ precipitate had formed in almost all these samples. Apparently enough air had diffused into the bottles to oxidize the ferrous ion. Because Fe(OH)$_3$ is a good scavenger for plutonium, we filtered these suspensions through 0.05-µm membrane filters and determined the filtrate plutonium concentrations. The results, shown in Table 3, indicate that, in a number of the trench leachates, an appreciable fraction of the plutonium is so stabilized in solution that it can withstand Fe(OH)$_3$ scavenging. Such resistance to carrier precipitation can be adequately explained only by the presence of strong complexes, such as those of EDTA, DTPA (diethylenetriamine pentaacetic acid), and NTA (nitrilotriacetic acid). Samples of the filtrates from the Fe(OH)$_3$ precipitations were analyzed for these three ligands, and a number were found to contain EDTA (Table 3). (No DTPA or NTA was detected.) However, when the supernates were fractionated on a Sephadex G-10 gel filtration column, it was found that the plutonium was distributed through several widely separated fractions. The gel filtration results are difficult to interpret, and it has not been possible to identify the organic ligands complexing plutonium. Workers at Brookhaven National Laboratory (*8*) have identified a number of organic compounds in Maxey Flats leachates, but none of these compounds—straight-chain and cyclic carboxylic acids, phenols, and alcohols—form complexes sufficiently stable to account for the behavior of plutonium. However, the procedure they employed (physical adsorption and solvent extraction followed by gas chromatography–mass spectrometry) would not separate highly polar, hydrophilic compounds such as EDTA, which form the most stable complexes. It is likely that some, but not all, of the plutonium is complexed by EDTA in most of the trench leachates. This behavior is similar to that observed by Means *et al.* (*9*), who found that EDTA in trench waters at a waste disposal site at Oak Ridge National Laboratory was responsible for the mobilization of cobalt.

Several conclusions can be drawn from these data. Except for leachate from trench 27, most of the plutonium is in true solution, in the tetravalent state, and exists in complexes with various organic ligands, including EDTA. Trench 27 leachate appears to contain an appreciable amount of colloidal plutonium, and little if any of the fraction in true solution is in the tetravalent state. Since the plutonium was carried by PrF$_3$, trench 27 leachate contains trivalent plutonium, as an organic complex, in addition to possible colloidal material. Oxidation state data for trench 33-L-4 leachate indicate that plutonium is predominantly tetravalent, but the presence of oxidized states is suggested by the PrF$_3$ data and is consistent with the high *p*H of this leachate. Considering the wide variety of waste materials in the trenches, it is somewhat surprising that the behavior of plutonium in their leachates is so similar.

The findings of this study have a bearing on the overall problem of ground water transport of plutonium and other actinides. Maxey Flats leachates are certainly not typical ground waters, but they may well be representative of the waters in shallow burial sites in areas of abundant rainfall. Our results indicate that in the presence of water containing strongly complexing organic species, plutonium can be mobilized to a significant degree as a soluble organic complex that can withstand submicrometer filtration, Fe(OH)$_3$ carrier precipitation, and surface adsorption on sediments. [Means *et al.* (*9*) have described the drastic decrease in K_d values for trace metals in the presence of even very small concentrations of strong complexing agents.] Whether these complexes existed in the waste at the time of disposal or were formed in the trenches after disposal is not known, but it is likely that there are some complexes from both sources. Hence it is important that all organic matter in transuranium wastes be destroyed in order to prevent the formation of stable, potentially mobile complexes of plutonium. Moreover, ground water in the area should be free of strongly complexing ligands. For this reason, it is highly inadvisable to locate a chemical waste disposal site adjacent to a radioactive waste disposal site. Naturally occurring organic substances in ground water appear less likely to mobilize plutonium than organic matter in the wastes. Results of an earlier study (*10*) indicate that plutonium is not appreciably solubilized by fulvic compounds in natural waters.

Although the results of this study indicate that the plutonium is in true solution, a previous investigation (*11*) of plutonium in a pond water indicated that the solubilized plutonium was predominantly colloidal. Hence it is clear that the chemical and physical form of plutonium, and therefore its migration behavior, varies widely with the composition of the water. Subsequent studies with different types of ground water should establish a more precise relation between plutonium speciation and ground water composition.

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Organic Fluorine in Human Serum: Natural Versus Industrial Sources

Abstract. *The concentration of organic fluorine in human serum has been reported to vary from 0.0 to 0.13 part per million in persons not exposed to industrial fluorochemicals. To help ascertain whether the natural environment is a source of organic fluorine in human serum, samples from a group of rural Chinese were analyzed. The samples contained low levels of organic fluorine as well as the expected inorganic fluoride.*

It has been observed (*1*) that there are two forms of fluorine in human serum: exchangeable and nonexchangeable with $^{18}F^-$. Research (*2*) has been directed toward identifying the nonexchangeable or organic form. Organic fluorine has been detected in human plasma (*3*); in one study it was not detected in the serum of a variety of animals (*4*), although in other studies it was found in bovine (*5*) and rat (*6*) serum. Since it has been suggested that organic fluorocompounds in human blood are derived from commercial sources (*2*), we analyzed serum from humans who live in a rural area in the People's Republic of China and compared the results with reported values for people in urban areas of the United States.

Several methods have been used to measure total fluorine in serum and plasma. Most samples have been analyzed by open ashing, which causes a variable loss of fluorine (*5*). For example, only 21 percent of the fluorine in perfluorooctanoic acid is recovered as inorganic fluoride by this method (*2*). The use of the closed oxygen bomb technique (*5–7*) avoids most of these losses. With this technique, recovery of fluorine from perfluorooctanoic acid is > 90 percent (*7*). The method yields a mean blank of 0.02 μg, which corresponds to 0.002 part per million (ppm) in a 10-ml serum sample.

Table 1. Concentrations of organic fluorine and inorganic fluoride in eight rural Chinese.

Person	Organic fluorine (ppm)	Inorganic fluoride (ppm)
1	0.008	0.051
2	0.013	0.054
3	0.011	0.046
4	0.014	0.046
5	0.009	0.044
6	0.009	0.049
7	0.004	0.046
8	0.017	0.076

Eight samples of human serum were obtained from Chinese donors who live in a rural commune, with little chance for exposure to industrial fluorochemicals. The samples were analyzed for organic fluorine and inorganic fluoride (F^-) by the oxygen bomb method. As shown in Table 1, all the samples from the Chinese contained detectable concentrations of organic fluorine. These concentrations are at the low end of the range compared to those in groups representing a more urban environment. Ash analysis of 65 plasma samples from residents of New York State gave an average value for organic fluorine of 0.03 ppm (lowest value, 0.005 ppm) (*3*). In plasma samples from 106 individuals living in five cities in two states, a mean organic fluorine concentration of 0.025 ppm (ashing) was observed, with two samples estimated to contain < 0.005 ppm (*2*). In plasma samples from 264 people in one Minnesota community, the average concentration of organic fluorine was 0.045 ppm (ashing); one sample contained no detectable organic fluorine (0.00 ppm) (*8*). Ash analysis of a pooled serum sample from Argentinians showed an organic fluorine concentration of 0.085 ppm (*9*). Oxygen bomb analysis of serum samples from nine Minnesota residents gave an average value of 0.02 ppm (lowest value, 0.01 ppm) (*7*).

The concentrations of F^- in the Chinese were slightly higher than the 0.02 ppm reported by Belisle and Hagen (*7*)